LA-UR-21-28089
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Intended for: Report

Issued: 2021-08-12

Improved temperature control for measuring the humidity dependence of aerosol optical properties

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ABSTRACT

Aerosols impact climate and cloud formation by their ability to take up water, which leads to changes in their optical properties. To better characterize the interactions of aerosol with water vapor, a humidification section has been added to the Cavity Attenuation Phase-shift Spectrometry (CAPS) instrument. The CAPS is a diode based method that provides simultaneous measurements of both aerosol total optical extinction and scattering in real-time. We are improving the humidity controlled sample line with new temperature controls. The current system suffers from particle-losses and poorly controlled humidity profiles, both of which were corrected for in the systems redesign. This improved wet-CAPS system has been integrated with a dry-CAPS to simultaneously measure the wet and dry aerosol optical properties. The two CAPS monitors were operated in a dry collection mode to ensure there were no biases between the instruments. Initial calibrations and data were collected to test the functionality of the dual CAPS system (wet + dry). The new two channel CAPS system improves our ability to measure the effects of humidity on aerosol optical properties.

I. INTRODUCTION

Aerosols are fine solid or liquid particles suspended in the atmosphere and are generally comprised of inorganic or organic substances created by natural processes (forest fires, desert dust, and sea spray) or human activities (burning of fossil fuels, bio fuels, and vehicle combustion). They can reside within the Earth's atmosphere for days or weeks before falling to the ground or being washed out by rain or snow[1]. In Earth's atmosphere, the aerosol particles are exposed to weather conditions that change their physical properties and affect their interactions with solar radiation. These changes are important to study and understand for human health, climate change, and cloud formation.[2]

A. Optical Properties of Aerosols

Solar radiation in the form of sunlight can interact with atmospheric aerosols by colliding with them. When this happens, the energy from the light is either scattered or absorbed by the aerosol. The total effect of scattering and absorption is referred to as extinction. (Figure 1).

Light scattering is when an electromagnetic wave (light) is forced to deviate from a straight trajectory by an obstacle (aerosol). As the electromagnetic wave interacts with the discrete particle, the electron orbits within the particle's constituent molecules are perturbed periodically with the same frequency as the electric field of the incident wave. The oscillation or perturbation of the electron cloud results in a periodic separation of charge within the molecule which is called an induced dipole moment. The oscillation induced dipole moment is manifest as a source of electromagnetic radiation, thereby resulting in scattered light[3].

Models of light scattering can be divided into three domains. If the particle is smaller than the wavelength of light, Rayleigh scattering is used. If the particle is about the same size as the wavelength of light, Mie scattering is used. If the particle is much larger than the wavelength of light, geometric scattering is used. As the particles grow larger, the complexity of the calculations grow[3].

Light absorption is when an electromagnetic wave (light) transfers it's energy into internal energy of the obstacle (particle). The absorbed energy can increase the charged particle's kinetic energy, potential energy, or both, leading to effects such as heating and atomic transitions. The absorbed momentum can change the particle's motion, leading to effects such as ionization and

radiation pressure[4].

From light scattering and absorption, we can calculate two other optical properties. Light extinction is the loss of light in the atmosphere from a directly transmitted beam due to the combination of light scattering and absorption.[5]. Aerosol single scattering albedo is defined as the ratio of the aerosol scattering coefficient and the extinction coefficient which is the sum of the absorption and scattering coefficients[6].

Light can also be refracted and is measured by the refractive index. The ratio of the speed of light in a vacuum to that in a second medium of greater density[7].

B. Humidified Aerosols and Water Uptake

The aerosols within the atmosphere can be very diverse due to their origins, causing complexities in their chemical properties and how they react with relative humidity (RH). In very general terms, we can classify the organic compounds as being hydrophobic[8] and the inorganic compounds as being hydrophilic[9]. This classification reflects how much water uptake occurs when RH is present in the atmosphere. In an inorganic aerosol, the amount of RH is proportional to the amount of water uptake. In an organic aerosol, lower amounts of RH may not experience any water uptake while in higher amounts of RH the aerosol might only experience a slight water uptake[10]. The amount of water uptake by an inorganic or organic aerosol can change its lifetime, particle cross-sectional area, volume, composition, and their refractive index[10]. Therefore, changing the aerosol's optical properties. (Figure 2).

C. Relative Humidity and Temperature

Relative humidity (RH) can be expressed as the ratio of the partial pressure of water vapor in an air-water mixture to the equilibrium vapor pressure of water over a flat surface of pure water at a given temperature[11].

The temperature of the water affects RH by changing the partial pressure of water vapor. As the temperature of water increases, the kinetic energy of its molecules increases and the number of molecules transitioning into water vapor also increases, thereby increasing the vapor pressure and the RH[12]. (Figure 3).

II. METHODS

In order to understand how humidity dependency affects the optical properties of aerosols, we need to simulate different levels of humidity to characterize the roll of water uptake on their optical properties. This can be done by building a system to expose aerosol samples to different levels of RHs by changing the water temperature and therefore the partial pressure of the water vapor of the sample air stream. This results in a humidified sample that passes to an instrument to measure its aerosol optical properties. A second instrument will be used to measure the optical properties of ambient aerosols at ambient RH and used as a reference for data analysis.

A. Instruments for Measuring Optical Properties

Measuring optical properties of humidified aerosols has been done before in previous scientific papers and several techniques have been developed using different methods and instrumentation.

The cavity-attenuated phase shift-single scattering albedo particulate matter monitor (CAPS PM_{ssa}; Aerodyne Research, Inc., Billerica, MA, USA) is an instrument used for simultaneous measurements of both particle-based total optical extinction and scattering and produces a direct measurement of the particle single scattering albedo, the direct ratio of scattering to total extinction. It uses a diode with a wavelength set to 450nm (blue light)[13].

The aethalometer (Magee Scientific, CA, USA) continuously collects and analyzes aerosol particles through an aerosol laden air stream that is drawn through a spot on a filter tape set at a measured flow rate. Simultaneously illuminated by light transmitted through an unexposed portion of the tape, acting like a reference. Optically absorbing material accumulates on the spot as the intensity of light is transmitted and gradually decreases. The decrease in light intensity from one measurement to the next is interpreted as an increase in collected material and the increase amount is divided by the known air flow volume to calculate the concentration[14].

The photo acoustic soot spectrometer (PASS, Droplet Measurement Technologies, CO, USA) is a three wavelength instrument that measures the optical absorption coefficient of suspended aerosol particles using a variation of the photo acoustic technique which is used to measure optical absorption of gases. This can be converted to black carbon mass loading based on an assumed specific absorption of black carbon to calculate the result[15].

The particle soot absorption photometer (PSAP, Brookhaven National Laboratory, NY, USA)

is used to measure in near real-time the light absorption coefficient based on the integrating plate technique in which the change in optical transmission of a filter caused by particle deposition on the filter is related to the light absorption coefficient of the deposited particles using Beer's Law[16].

Out of these instruments, the aethalometer and PSAP are filter based and measure deposits rather than in situ aerosol[17]. The filter paper itself isn't always optically consistent and is subject to being overloaded by an excess amount of aerosols. The filter absorption overestimates the results from imperfect scattering corrections[18] and organic compounds modify the substrate optical properties[19]. The filter affects the water uptake, leaving these two instruments unsuitable for RH studies[20].

The CAPS PM_{ssa} and PASS instruments use in situ techniques for measurements. The PASS is subject to interference at higher RH due to laser induced evaporation, limiting its utility[2122]. The CAPS PM_{ssa} measures single-scattering albedo (SSA) and quantifies the relative importance of scattering particles at a given wavelength and the SSA depends on RH making it the logical choice for this application. However it does have one major disadvantage, the wavelength is fixed at 450 nm blue light.

B. Temperature Control System to Humidify Aerosols

The new humidification system uses a TC-48-20 temperature controller (TE Technology, MI, USA) paired with a MP-3193 thermistor (YSI Inc, OH, USA) and a LC-066SS liquid cooler (TE Technology, MI, USA). The thermistor sends the water temperature of the system ($^{\circ}C$) to the temperature controller which then tells the liquid cooler to adjust the water temperature accordingly (Figure 4). The humidifier is designed with two cylindrical tubes with a water and aerosol intake and outtake. The sampled aerosols enter through the small inner tube surrounded by a diffusion membrane.

A new water pump and reservoir move the water through the closed loop system. The water enters through the large outer tube and passes through the diffusion membrane to humidify the aerosol sample. The water exits back into it's closed system and the aerosols exit through plumbing to the CAPS Wet system for measurement (Figure 5). Both the sample aerosol and water lines and connections are wrapped in insulated tape to maintain a constant temperature within the individual systems.

III. RESULTS

A. Measurements Dual-CAPS comparison

In order to test the two CAPS instruments side by side, a sample of Fullerene Soot, a black carbon surrogate was measured under ambient conditions without humidification. The data from this test was visualized using a direct comparison graph and box plot in order to determine the slope, median, and percent error between the two systems.

In Figure 6 (Direct Comparison), the measurements from both machines are directly compared for extinction and scattering with a linear line fit (red) to calculate the slope. In Figure 7 (Distribution of the Difference), the difference is calculated by subtracting the measurements from CAPS2 by CAPS1. The result is then graphed on a box plot to calculate the median of the difference between the two points. The percentage error is calculated by taking the difference and dividing it by the average of the two measurements. The resulting column is then averaged for both extinction and scattering.

Looking at the results from the table, the slope is very close to a one-to-one line, the median of the difference is close to zero, and the percent error is less than 1%. This shows a slight bias between the measurements of the two machines but is fairly marginal. With this bias in mind, we can be confident in our ability to compare the wet and dry CAPS measurements.

Property	**Slope**	**Median**	**Percentage Error**
Extinction	1.054	0.0524	0.7%
Scattering	1.072	0.0607	0.9%

TABLE I: Results from CAPS comparison.

IV. FUTURE WORK

Now that the temperature control and humidification systems are set up with a designated dry and wet line for the two CAPS instruments we can test the measurements from the dry line against the measurements from the wet line. We still need to determine if there is an improvement in

the temp/RH control within the system. From there we can start running tests on different absorbing and non-absorbing aerosols to further explore their optical properties under ambient and humidified conditions.

V. ACKNOWLEDGEMENTS

This work was supported in part by the U.S. Department of Energy, Office of Science, Office of Workforce Development for Teachers and Scientists (WDTS) under the Community College Internships (CCI) program.

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VI. APPENDIX

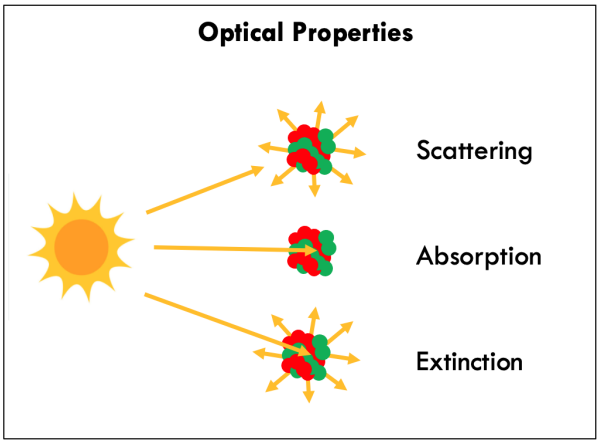

FIG. 1: Optical Properties of Aerosols. Sunlight interacts with aerosol particles in the form of scattering, absorption, and extinction.

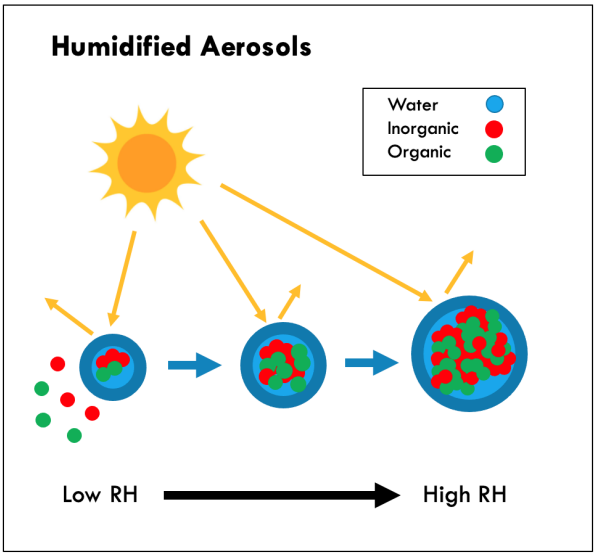

FIG. 2: Humidified Aerosols and Water Uptake. As the relative humidity increases, the aerosol properties change.

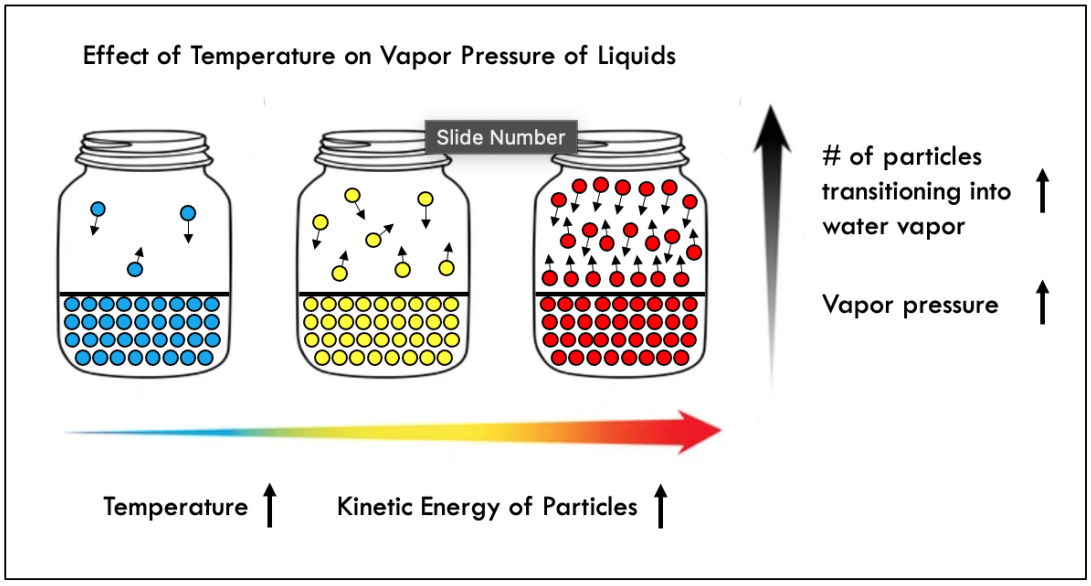

FIG. 3: Relationship between vapor pressure and temperature.

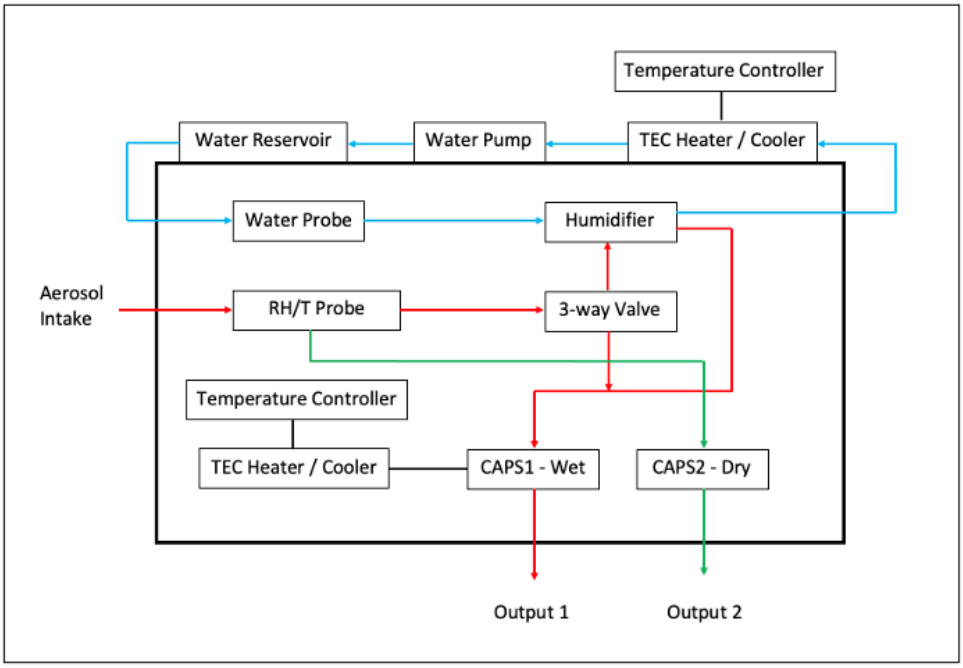

FIG. 4: Dual CAPS Design. The blue line represents the water flow, the red line is the humidified aerosol sample line, and the green line is the ambient sampling line.

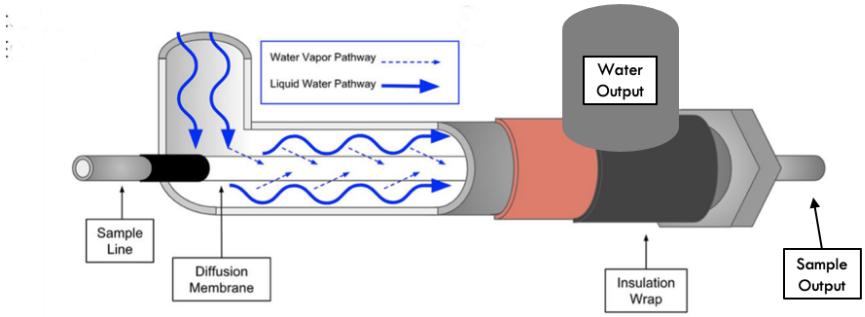

FIG. 5: Aerosol Humidifier. Aerosol and water intake with the flow direction and diffusion membrane. With the water and aerosol outtake. Wrapped in insulated tape.

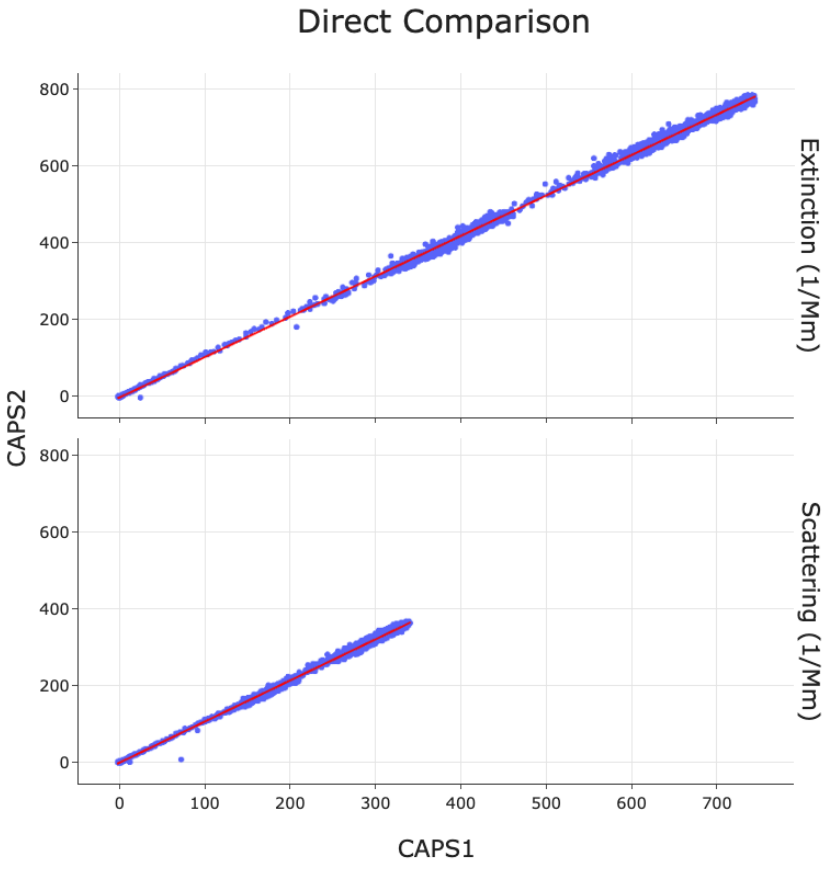

FIG. 6: Direct comparison. Plots the measurements from CAPS1 and CAPS2 for both extinction and scattering. The red line represents the line fit to calculate the slope.

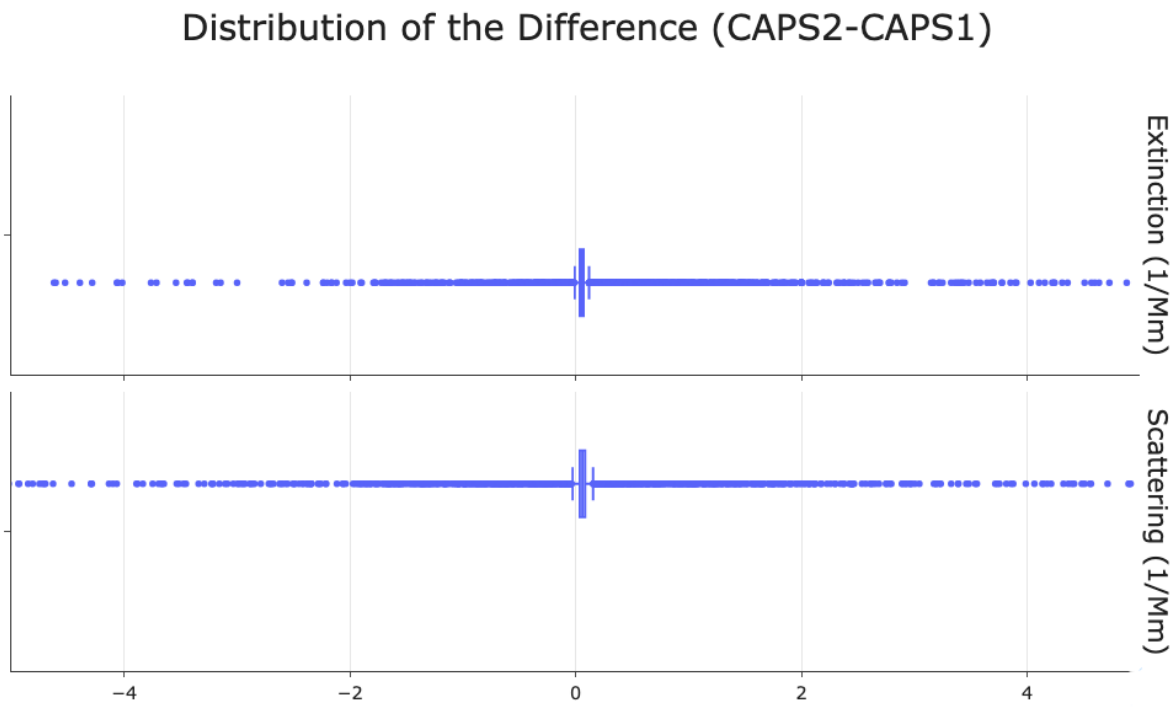

FIG. 7: Distribution of the Difference. The difference of the two CAPS measurements are plotted on a box plot to display the median of the difference.